METHOD AND APPARATUS FOR HEAT TREATMENT OF PARTICULATES IN AN ELECTROTHERMAL FLUIDIZED BED FURNACE AND RESULTANT PRODUCTS

Inventor: Mark S. Zak
William Mark Harrison
Joseph E. Doninger

BACKGROUND OF THE INVENTION

[0001] The present invention relates to a method and apparatus for the continuous high-temperature treatment in an electrothermally heated fluidized bed of carbonaceous particles comprising fine or irregularly shaped particles having a wide range of particle size distribution and the products resulting from such treatment. More particularly, the invention relates to, in one aspect, the use of a fountain-type fluidized bed for the high temperature treatment of carbonaceous particles that

cannot be effectively treated in a bubble-type fluidized bed due to their fine sizes, range of size distribution, and shape.

[0002] It is known to use an electrothermal fluidized bed (EFB) furnace for the high temperature purification of carbonaceous materials and for high temperature chemical synthesis (see U.S. Patent Nos. 4,160,813 and 4,547,430, respectively).

[0003] These processes use a fluidized bed furnace, as illustrated in U.S. Patent No. 4,543,240, in which the cross-section of the fluidized bed portion (or "fluidizing zone") of the EFB furnace is substantially constant along its height and the fluidizing gas is introduced into the furnace through a multiplicity of generally vertically oriented gas nozzles extending through a plate distributor at the bottom of the furnace. This type of EFB furnace is commonly referred to as a "bubble" EFB furnace.

[0004] The methods of purification and chemical synthesis using a bubble EFB furnace have worked well for particles as small as 106 µm (140 mesh). However, bubble EFB furnaces have not performed well with respect to smaller particles, particularly with those smaller than 75 µm (200 mesh). Additionally, such furnaces are not effective for use with irregularly shaped particles such as flakes and needles, and or

with particles having a wide range of particle-size distribution ("polydispersed"), particularly where the material comprises a high content (greater than 30%) of fine particles with sizes less than 106 µm (140 mesh).

[0005] The use of bubble EFB furnaces to treat and/or synthesize polydispersed materials has resulted in the entrainment of particles smaller than 106 µm (140 mesh). That is, the particles are entrained by the fluidizing gas outside of the active area of the EFB furnace. This results in a low recovery rate of treated product as a percent of raw material. This has proven to be especially the case in bubble EFB furnaces where the raw materials are introduced at the top of the fluidized bed and the treated particles are discharged from the bottom of the furnace.

[0006] With respect to fine particles, particularly those smaller than 45 µm (325 mesh), and those of irregular shape, it has proven very difficult, or at times impossible, to uniformly fluidize such particles in a bubble EFB furnace, because of channels of fluidizing gas. This is believed to be due to the high adhesion forces between the small particles that result from the relatively large surface area for fine particles and also because of stagnation zones formed in the bottom portion of the fluidized bed.

[0007] These shortcomings are the result of the particular hydrodynamics of a bubble EFB furnace. In particular, the plate gas distributor and its plurality of vertically oriented gas nozzles create a number of local circulating zones that have an upward flow of particle/gas mixture and a downward flow of particles, with each zone being formed around a single nozzle or group of nozzles on the distribution plate.

[0008] Accordingly, it is an object of the present invention to provide a method for treating of fine, irregularly shaped and/or polydispersed particulate matter in an electrothermal fluidized bed furnace. It is a related object to provide a furnace for performing the method.

SUMMARY OF THE INVENTION

[0009] These objects, and others which will become apparent upon reference to the following detailed description and drawing, are provided by an electrothermal fluidized bed furnace in which the furnace body has upper and lower cylindrical portions, with the upper cylindrical portion having a diameter larger than that of the lower cylindrical portion. A conical portion is disposed below the lower cylindrical portion so that the conical portion and the lower cylindrical portion define a fluidizing zone while the upper cylindrical portion defines an overbed zone. The furnace includes at least one electrode

extending through the upper and lower cylindrical portions and a treated material discharge pipe at the lower end of the conical portion. A feed pipe is provided for introducing raw material into the lower cylindrical portion, and at least one gas flue is provided at the top of the furnace body for discharging fluidizing gas. A plurality of nozzles is disposed in the conical section for introducing fluidizing gas into the furnace, with the nozzles being arranged in a generally horizontal plan and orientated so that the streams of the fluidizing gas introduced therethrough cross and form an upward flow in the central portion furnace body.

[00010] Such an electrothermal fluidized bed furnace is adapted to be used in a process for continuously heat treating particulate matter by continuously introducing a non-reactive fluidizing gas through the nozzles of the furnace at pre-determined rate, continuously introducing untreated particulate matter through the feed pipe of the furnace at a predetermined rate so that it forms a fluidized bed, energizing the electrode so as to heat the fluidized bed, and continuously collecting the treated particulate matter from the discharge pipe. Starting materials for the process advantageously include various types of cokes (e.g., fluid coke, flexi-coke, pitch coke, delayed coke and needle coke) and graphite materials (e.g., flake

graphite, synthetic graphite, amorphous graphite, and vein graphite).

BRIEF DESCRIPTION OF THE DRAWINGS

[00011] Fig. 1 is a vertical cross-sectional view of a fountain EFB furnace according to the present invention.

[00012] Fig. 2 is a top view of the fountain EFB furnace of Fig. 1.

[00013] Fig. 3 is a cross-sectional view of the EFB furnace taken along line 3-3 of Fig. 1, showing the fluidizing gas distribution nozzles.

[00014] Fig. 4 is similar to Fig. 3, except that it shows an alternative arrangement for the fluidizing gas distribution nozzles.

DESCRIPTION OF THE PREFERRED EMOBODIMENT

[00015] Turning to the figures of the drawings, there is seen a fountain-type EFB furnace, generally designated 10, in accordance with the present invention. The principal characteristic of a fountain fluidized bed (also known as a "spout" or "jetting" fluidized bed) is that it has a strong circulating contour with a central upward flow of particle-gas mixture in the center of the fluidized bed and an outer downward flow of particles along the furnace walls. The high speed central upward flow draws in and carries along the solid

particles. The formation of fine particle clusters and gas channels in the fluidized bed is avoided. The vertical velocity gradient provides for a thorough fluidization of all fractions of poly-dispersed grain materials.

[00016] With reference to Fig. 1, the furnace 10 includes a furnace shell 11, typically made of steel that encases a furnace body 12. If the operation temperature of the furnace is greater than 1500°C, made of graphite and constitutes the return electrode. The furnace body may be made of other materials if the operation temperature is less than 1500°C. An insulating material 14 is disposed between the shell 11 and body 12. The furnace body 12 comprises a lower cylindrical portion 16, an upper cylindrical portion 18 disposed above the lower cylindrical portion and having a larger diameter than the central cylindrical portion 16. (For the purposes of the description of the furnace 10, the term "cylindrical" means having vertical wall(s) and a constant cross section throughout its height.) A conical gas distributor 20 is disposed below the central cylindrical portion 16, and has a plurality of fluidizing gas distribution nozzles 22. The nozzles 22 are in fluid communication with a plenum 24 into which the fluidizing gas is introduced through an inlet 26. The conical gas distributor 20 defines a central angle α (alpha) of from 30° to

90°, and preferably of from 40° to 60°. In such a furnace body 12, the space above the gas distribution nozzles 22 to the top of the lower cylindrical portion 16 generally define the fluidized bed zone 28. The space above the fluidized bed zone, coinciding generally with the upper cylindrical portion 18, is known as the overbed space or free board zone 30. In the furnace of the present invention, the operational height H_{FB} of the fluidized bed area 28 generally coincides with the distance between the nozzles 22 and the upper end of the lower cylindrical portion 18. In order to prevent the formation of a bubble fluidized area in the top portion of the fluidized bed zone 28, H_{FB} is preferably less than or equal to one and one-half to twice the inside diameter ID_{FB} of the lower cylindrical portion 16. The minimal height of the free board or overbed space $H_{ov.s}$ is preferably one and one-half times the height of the fluidized bed H_{fb} to ensure that any entrained particles are separated from the gas flow and returned to the fluidized bed space of the furnace.

[00017] Preferably, each of the cylindrical portions 16, 18 and the conical gas distributor 20 has a circular or an elliptical cross-section. Other shaped cross-sections (such as square, rectangular, octagonal, etc.) may exhibit satisfactory hydro-dynamic characteristics. However, such shapes are

practically unworkable due to the amount of thermal expansion encountered by the furnace during use.

[00018] An elongated electrode 32 extends into the furnace body 12 from the top 34 through the upper and lower cylindrical portions 18, 16, respectively. The electrode 32 is preferably fabricated from an electrically conductive, heat-resistant material such as graphite and must be electrically isolated from furnace body 12. When a single electrode is used, it must be located centrally within the furnace body and aligned with a vertical axis Y thereof. Alternatively, a plurality of electrodes may be used, in which case the electrodes are arranged symmetrically about the central axis Y.

[00019] A feed pipe 38 is provided for continuously supplying raw material into the fluidized bed zone 28 of the furnace body 12. As illustrated, the feed pipe 38 is vertically orientated and extends through the top 34 of the furnace body 12, down through the upper cylindrical portion 18, and has its outlet adjacent to the wall either at or below the top of the lower cylindrical portion 16. As such, raw material is introduced from the feed pipe 38 into the fluidized bed, or at least at the top surface thereof, in the area of the downward flow of solid particles being circulated in the fluidized bed. This results in easier loading of raw material into the fluidized bed, reduces

the likelihood of the untreated particles being entrained by the upward flow of fluidizing gas and carried into the overbed space, and provides better mixing of the treated and raw materials.

[00020] The bottom of the furnace body includes a discharge port 40 through which effluent solids may be continuously withdrawn by gravity flow. The discharge port 40 depends from the conical gas distributor 20, with the inlet to the discharge port 40 generally coinciding with the apex of the conical gas distributor 20.

[00021] Gaseous effluent can be withdrawn through one or more exhaust pipes or gas flues 42 in the top 34 of the furnace body 12. This effluent gas can be readily cleaned and treated to control particulate and gaseous pollutants as required.

[00022] In keeping with the invention, the conical gas distributor 20 includes a plurality of fluidizing gas inlet nozzles 22 (eight shown), through which fluidizing gas is introduced into the furnace body 12. The nozzles 22 are orientated radially to the center of the conical distributor 20 so that fluidizing gas forms crossing sprays, with a strong uniform upward flow. As can be appreciated, the velocity at which the fluidizing gas exits the nozzles and the average gas velocity in the fluidized bed portion 16 depend on the particle

size, density, and shape of the material being fluidized. In the context of the process of the present invention, the fluidizing gas is typically nitrogen, argon or other non-reactive gas.

[00023] In one embodiment, best seen in Fig. 3, the nozzles 22 are arranged so that their axes X are aligned radially, with the fluidizing gas being directed toward the center of the conical gas distributor 20. Alternatively, the nozzles 22 maybe orientated so that their axes X form an angle β of from 10 to 20° with respect to a tangent to the conical gas distributor 20 at the location of the nozzle, as best seen in Fig. 4. The arrangement of the nozzles 22 so that their axes X are generally tangential to nozzle circle provides for a rotation of the fluidized bed, making it more stable and less sensitive to any deviation of the elongated electrode 32 from the central axis Y. This angle helps to prevent the fluidized particles from being brought into contact with the conical gas distributor 20 at high velocity, which could result in undue wear of the walls of the gas distributor 20 from abrasion.

[00024] In order to prevent the fluidizing gas from interfering or disrupting the discharge of treated particles from the furnace 10, the nozzles 22 are preferably disposed at a height H_N above the conjunction of the gas distributor 20 and the

inlet to the discharge port 40. Preferably, H_N is from 0.5 to 0.75 of the total height H_{TC} of the conical gas distributor 20, and more preferably from 0.6 to 0.65 H_{TC}.

[00025] Each of the nozzles 22 has preferably a ring cross section perpendicular to its X axis at which is defined a free cross-sectional area. The cross-sectional shape can be circular or can have another shape such as rectangular, oval etc. The sum of the free cross-sectional areas of the nozzles 22 should be from 0.15 to 0.5% of the cross-sectional area of the cylindrical portion of the fluidized bed, that is the cross-sectional area of the lower cylindrical portion 16. Preferably, the free cross-sectional area of the nozzles 22 should be between 0.25 and 0.4% of the cross-sectional area of the fluidized bed.

[00026] From the foregoing, the method for treating fine particulate materials in the inventive EFB furnace should be self-evident. First, untreated particulate material is continuously fed by gravity through the feed pipe 38 into the reaction zone of the EFB furnace 10. The untreated particulate material may comprise fine, irregularly shaped or polydispersed materials. In pilot runs, the polydispersed material has comprised particles sized from between 1.7mm (12 mesh), and as small as 5μm. Further, the untreated particulate may be an

electroconductive or semiconductive material, such as carbonaceous materials like carbon black, coke (fluid coke, flexi-coke, delayed coke, needle coke, pitch coke, etc.), and graphite (flake graphite, synthetic graphite, vein graphite, amorphous graphite, etc.). The various cokes may be either green or calcined, petroleum or metallurgical, and are widely available from various sources. The graphites are available from Superior Graphite Co. of Chicago, Illinois, the assignee of the present application. The untreated particulate matter is discharged from the feed pipe 38 at the top of, or just inside, the fluidizing zone in the downward flow of particles.

[00027] The material from the feed pipe is maintained in a fluidized state in the region of the furnace corresponding approximately to the lower cylindrical portion 16, and electric current is passed through the fluidized bed to uniformly heat the material to a high temperature, typically from 2,200-2,400°C.

[00028] Treated particulate material is continuously withdrawn by gravity through the discharge pipe 40. The discharge rate is such that the treatment time of the particulate material within the fluidized bed is sufficient to result in the desired heat treatment or chemical reaction. In the use of the present EFB furnace, there is no need for mechanical devices or moving parts

within the furnace 10.

[00029] After being discharged through the pipe 40, the treated material may be cooled in a cooling chamber (not shown). Gaseous effluent can be withdrawn through the gas flue 42 at the top 34 of the furnace body 12. This gaseous effluent can be readily cleaned and treated to control pollutants to the extent required.

[00030] By use of the inventive EFB furnace and the heat treatment fine particles, significantly better recovery rates (of 90.3% in pilot runs) for treated particulate have resulted, in contrast to the recovery rates when using the prior art, bubble-type EFB's, (in which the recovery rate is typically less than 64%). In addition, the critical velocity of fluidization has been reduced over that of a bubble-type EFB furnace by 10-15%, for example from approximately 0.30 ft./sec. to approximately 0.25 ft./sec. in the inventive EFB furnace.

[00031] Table I below compares purity characteristics for several different graphite and coke materials both prior to and after heat treatment according to the present invention. The purity characteristics compared are percentage (wt.) of ash and sulfur.

TABLE I

Purity of Various Carbonaceous and Graphitic Thermally Processed Materials			
Material Description		**Ash Content %**	**Sulfur Content %**
Flake Graphite (Coarse)	Feed Treated	0.9-1.15 0	- -
Flake Graphite (Fine)	Feed Treated	1.5-1.65 0	0.03-0.04 0.0012
Fluid Coke	Feed Treated	0.6-0.7 0	1.9-2.0 0.007
Carbon (Pellets)	Feed Treated	- -	0.48 0.004

[00032] Thus, an improved EFB furnace and method for the treatment of fine particulates have been provided. While the invention has been described in terms of a preferred embodiment and method, there is no intent to limit it to the same. For example, the furnace and process is equally well suited for chemically treating find particulates, in which case the fluidizing gas can be a reducing gas, such as carbon monoxide, hydrogen, methane, etc. Instead, the invention is defined by the scope of the appended claims.